

TITLETHE USE OF TWO OR MORE SENSORS  
IN A NUCLEAR QUADRUPOLE RESONANCE DETECTION SYSTEM  
TO IMPROVE SIGNAL-TO-NOISE RATIO

5

This application claims the benefit of U.S.  
10 Provisional Application No. 60/541,660, filed on  
February 4, 2004, and U.S. Provisional Application No.  
60/557,235, filed on March 29, 2004, each of which is  
incorporated in its entirety as a part hereof for all  
purposes.

15

Field of the Invention

This invention relates to a nuclear quadrupole  
resonance detection system and to the use of two or  
20 more sensors tuned to the same nuclear quadrupole  
resonance frequency when detecting the nuclear  
quadrupole resonance signal, thereby providing improved  
nuclear quadrupole resonance detection system  
performance.

25

Background of the Invention

The use of nuclear quadrupole resonance (NQR) as a  
means of detecting explosives and other contraband has  
30 been recognized for some time. See e.g. T. Hirshfield  
*et al*, *J. Molec. Struct.* 58, 63 (1980); A.N. Garroway  
*et al*, *Proc. SPIE* 2092, 318 (1993); and A.N. Garroway  
*et al*, *IEEE Trans. on Geoscience and Remote Sensing* 39,  
1108 (2001).

35

NQR provides some distinct advantages over other  
detection methods. NQR requires no external magnet  
such as required by nuclear magnetic resonance. NQR is

sensitive to the compounds of interest, i.e. there is a specificity of the NQR frequencies.

5 One technique for measuring NQR in a sample is to place the sample within a solenoid coil that surrounds the sample. The coil provides a radio frequency (RF) magnetic field that excites the quadrupole nuclei in the sample and results in their producing their characteristic resonance signals. This is the typical  
10 apparatus configuration that might be used for scanning mail, baggage or luggage.

There is also need for a NQR detector that permits detection of NQR signals from a source outside the  
15 detector, e.g. a wand detector, that could be passed over persons or containers as is done with existing metal detectors. Problems associated with such detectors using conventional systems are the decrease in detectability with distance from the detector coil  
20 and the associated equipment needed to operate the system.

A detection system can have one or more devices (such as coils) that both transmit and receive, or it  
25 can have separate devices (such as coils) that only transmit and only receive. A transmit, or transmit and receive, coil of an NQR detection system provides a magnetic field that excites the quadrupole nuclei in the sample and results in their producing their  
30 characteristic resonance signals that the coil receives. As the NQR signals have low intensity and short duration, the transmit, receive, or transmit and receive, coil preferably has a high quality factor (Q). The transmit, receive, or transmit and receive, coil  
35 has typically been a copper coil and therefore has a Q of about  $10^2$ .

It can be advantageous to use a transmit, receive, or transmit and receive coil, made of a high temperature superconductor (HTS) rather than copper since the HTS self-resonant coil has a Q of the order of  $10^3$ - $10^6$ . The large Q of the HTS self-resonant coil produces large magnetic field strengths during the RF transmit pulse and does so at lower RF power levels. This dramatically reduces the amount of transmitted power required to produce NQR signals for detection, and thereby reduces the size of the RF power supply sufficiently so that it can be run on portable batteries.

The large Q of the HTS self-resonant coil also plays an important role during the receive time. In view of the low intensity NQR signal, it is important to have a signal-to-noise ratio (S/N) as large as possible. The signal-to-noise ratio is proportional to the square root of Q so that the use of the HTS self-resonant coil results in an increase in S/N by a factor of 10-100 over that of the copper system.

These advantages during both the transmit and the receive times enable a detector configuration that is small and portable. In particular, the use of a high temperature superconductor sensor receive coil prepared from a high temperature superconductor material, provides a distinct advantage over the use of an ordinary conductor coil.

An object of the present invention is to provide a NQR detection system with improved performance, particularly where the transmit, receive, or transmit and receive coil, is made of a high temperature superconductor.

Summary of the Invention

One embodiment of this invention is a method of detecting nuclear quadrupole resonance in an object by

- a) providing two or more sensors tuned to a  
5 specified nuclear quadrupole resonance frequency, wherein each sensor receives the specified nuclear quadrupole resonance signal;
- b) applying a radio frequency magnetic field to the object; and
- 10 c) adding coherently the signals detected by the sensors.

Another embodiment of this invention is a nuclear quadrupole resonance detection system for detecting  
15 nuclear quadrupole resonance in an object, comprising:

- a) two or more sensors tuned to a specified nuclear quadrupole resonance frequency, wherein each sensor receives the specified nuclear quadrupole resonance signal; and
- 20 b) means to add coherently nuclear quadrupole resonance signals detected by the sensors.

A further embodiment of this invention is a nuclear quadrupole resonance detection system for  
25 detecting nuclear quadrupole resonance in an object, comprising:

- a) two or more sensors tuned to a specified nuclear quadrupole resonance frequency, wherein each sensor receives the specified nuclear quadrupole resonance signal; and
- 30 b) an electrical path from each sensor to a combination point at which the signals are added,

wherein the signals received across the paths add  
35 constructively at the combination point.

Preferably, the two or more sensors are used solely for sensing, i.e. receiving, the NQR signal, and

one or more separate coils are used as the transmit,  
i.e. excitation, coils to provide the RF magnetic field  
that excites the quadrupole nuclei in the object to be  
scanned. Preferably, the two or more sensors are high  
5 temperature superconductor coils. More preferably, the  
two or more sensors are each comprised of a high  
temperature superconductor self-resonant planar coil,  
or of two or more coupled high temperature  
superconductor self-resonant planar coils.

10

This invention for improving the signal-to-noise  
ratio and thereby the performance of a nuclear  
quadrupole resonance detection system is especially  
important when the nuclear quadrupole resonance  
15 detection system is used for detecting the nuclear  
quadrupole resonance of explosives, drugs and other  
contraband.

## 20 Brief Description of the Drawings

Figure 1 shows the HTS coil design of an HTS coil  
used in the example.

Figure 2 is a schematic diagram of the  
experimental set-up used in the example.

25

## Detailed Description of the Preferred Embodiments

This invention provides a method for increasing  
the signal-to-noise ratio of a NQR detection system for  
30 scanning a potential source of NQR, and also provides a  
NQR detection system that provides this increased  
performance. To accomplish this, two or more sensors  
are tuned to a specified nuclear quadrupole resonance  
frequency, and the signals detected by each sensor are  
35 added coherently. Two or more sensors are tuned to a  
"specified" nuclear quadrupole resonance frequency in  
the manner contemplated by this invention when each  
sensor is tuned to a frequency within a given nuclear

quadrupole resonance emission line so that even if each sensor is not tuned to exactly the same frequency, they are all still detecting the same emission line.

5           A "sensor" as used in this invention is a receive device, such as a receive coil, and the two or more sensors used to detect nuclear quadrupole resonance in a sample or object will typically be used as receive coils only. In certain embodiments, it may be possible  
10   to use them as transmit and receive coils, but, preferably, separate coils are used to transmit the RF signal, and the sensors (as their name implies) are used solely as receive coils.

15           The transmit coils used in this invention can be made of copper, silver, aluminum or a high temperature superconductor. A copper, silver or aluminum coil is preferably in the form of a shielded-loop resonator (SLR) coil. SLR's have been developed to eliminate the  
20   detuning effect of the electrical interaction between the coil and the surrounding material. Preferably, a copper SLR transmit coil is used to apply the RF signal to the sample or object to be scanned. In an NQR detection system, provision must of course be made for  
25   a power supply to supply power for transmitting the RF pulse.

          Preferably, the two or more sensors are high temperature superconductor (HTS) coils. A high  
30   temperature superconductor coil is preferably in the form of a self-resonant planar coil, i.e. a surface coil, with a coil configuration of HTS on one or both sides of a substrate. High temperature superconductors are those that superconduct above 77K. High  
35   temperature superconductors used to form a HTS self-resonant coil are preferably selected from the group consisting of  $\text{YBa}_2\text{Cu}_3\text{O}_7$ ,  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$ ,  $\text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_9$ ,  $(\text{TlPb})\text{Sr}_2\text{CaCu}_2\text{O}_7$  and  $(\text{TlPb})\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_9$ . Most preferably,

the high temperature superconductor is  $\text{YBa}_2\text{Cu}_3\text{O}_7$  or  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$ . A HTS self-resonant coil can be formed by various known techniques. A preferred technique for forming a  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  coil is described in the example.

It is often advantageous to be able to fine tune the resonance frequency of a sensor. One means for accomplishing such tuning is to use two or more coupled high temperature superconductor self-resonant coils. The resonance frequency of the fundamental symmetric mode of coupled high temperature superconductor self-resonant coils can be varied by mechanically displacing the coils with respect to one another, and such tunable coupled coils may be used as a HTS sensor. A coil used in a coupled configuration is preferably a planar, *i.e.* surface, coil. A planar coil may have a HTS coil configuration on only one side of the substrate, or may have essentially identical HTS coil configurations on both sides of the substrate. Most preferably, each sensor herein is comprised of a high temperature superconductor self-resonant planar coil, or two or more coupled high temperature superconductor self-resonant planar coils.

As indicated above, when two or more coupled high temperature superconductor self-resonant coils are used as a sensor, the resonance frequency of the fundamental symmetric mode of the coupled high temperature self-resonant coils can be varied by mechanically displacing the coils with respect to one another. Means for tuning the resonance frequency of a sensor to a specified nuclear quadrupole resonance frequency thus includes the mechanical displacement of coupled coils as described above.

Alternatively, for a sensor comprised of one or more high temperature superconductor self-resonant coil

coils, means for tuning the resonance frequency of the sensor to a specified nuclear quadrupole resonance frequency may include a circuit. The circuit can be comprised of a single loop or coil to inductively  
5 couple the circuit to the high temperature superconductor self-resonant sensor, a reactance in series with the single loop or coil, and means for connecting and disconnecting the reactance to and from the single loop or coil. In essence, the tuning  
10 circuit is resonated, which causes a mode split by coupling to the self-resonant high temperature superconductor sensor.

In the tuning circuit, the single loop or coil can  
15 be made of a regular conductor such as copper or a high temperature superconductor. The reactance can be an inductance, capacitance or combination of both. Means for connecting and disconnecting the reactance to and from the single loop or coil may include at least one  
20 mechanical switch, or electrical switch such as a diode. Preferably, the reactance can be varied so that the resonance frequency can be adjusted to more than one frequency. One way of providing a variable reactance is to utilize as the reactance two or more  
25 capacitors in parallel, each of which can be individually connected to or disconnected from the single loop or coil. Alternatively, a variable reactance can be prepared from two or more inductors in series, each of which can be individually connected to  
30 or disconnected from the single loop or coil by a mechanical or electrical switch that can short-circuit the inductor and thereby essentially remove it from the circuit.

35 The NQR signals detected by the two or more sensors used in this invention can be added coherently, i.e. the phases of the individual signals are, by various analog and digital techniques, adjusted to add



constructively. In the analog technique for coherent addition, the electrical path from each sensor to the combination point, at which the signals are added, is adjusted so that the signals add constructively at the combination point. When the sensors are essentially equidistant from the object that is the source of the nuclear quadrupole resonance signal, the electrical path from each sensor to the combination point can be made essentially identical, thereby insuring that the signals add constructively at the combination point.

Means for adding signals may also be provided, which may include a microprocessor for performing various digital techniques for coherent addition. In a digital technique, for example, the signal detected by each sensor may be multiplied before combination by a constant complex factor that can be measured or calculated to correct for phase differences between the signal paths and thereby insure that the signals add constructively at the combination point. The constant complex factor is specific to the electrical path from each sensor to the combination point. Typically, the signals will be amplified before they are added.

In this invention, the advantages in using in an NQR system an array of two or more sensors over the use of a single sensor can be seen as follows. The signal  $S$  obtained by using an array of  $n$  sensors and coherently adding the signals is proportional to  $n$ . Assuming that the noise present is random, the noise  $N$ , after the coherent addition, is proportional to  $n^{0.5}$ . Therefore  $S/N$  is proportional to  $n^{0.5}$ , i.e. is proportional to the square root of  $n$ . An array of two sensors therefore increases  $S/N$  by a factor of 1.4. An array of four sensors increases  $S/N$  by a factor of 2.

The use of HTS self-resonant coils as sensors makes the instant invention especially attractive. The

HTS self-resonant coils have high Q's and relatively small size and make the use of an array of two or more sensors more feasible. If one or more HTS coils are used herein as sensors, provision must be made for  
5 cooling the HTS to at least liquid nitrogen temperature.

The advantageous effects of this invention are demonstrated by an example, as described below. The  
10 embodiments of the invention on which the example is based are illustrative only, and do not limit the scope of the appended claims. The purpose of the example is to demonstrate the increase in S/N when two sensors are used to detect a frequency and the signals from the  
15 sensors are added coherently.

HTS self-resonant sensors are provided, each of which is two coupled essentially identical  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  planar coils. Each of the coupled coils is on a  
20 lanthanum aluminate ( $\text{LaAlO}_3$ ) substrate with the coil design configuration shown in Figure 1 on both sides of the substrate.

A clean, polished single crystal  $\text{LaAlO}_3$  substrate  
25 with a diameter of 2 inches (5.1 cm) and an approximate thickness of 0.02 inches (0.5 mm) was obtained from Litton Airtron, Morris Plains, NJ. Off-axis magnetron sputtering of a Ba:Ca:Cu oxide target is used to deposit, at room temperature (about 20°C), an amorphous  
30 precursor Ba:Ca:Cu oxide film on both sides of the substrate. This amorphous Ba:Ca:Cu oxide film is about 550 nm thick and had a stoichiometry of about 2:1:2. The precursor film is then thallinated by annealing it in air for about 45 minutes at 850°C in the presence of  
35 a powder mixture of  $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  and  $\text{Tl}_2\text{O}_3$ . When this powder mixture is heated,  $\text{Tl}_2\text{O}$  evolves from the powder mixture, diffuses to the precursor film and reacts with

it to form the  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  phase on both sides of the substrate.

5 The  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  is then coated with photoresist on both sides and baked. A coil design mask with the design shown in Figure 1 is prepared. The coil has an inner radius of about 10.5 mm and an outer radius of about 22.5 mm. The outermost HTS ring 31 of the coil is about 2 mm wide and the innermost HTS ring 32 is  
10 about 3.5 mm wide. The intermediate HTS rings 33 are about 250  $\mu\text{m}$  wide with about 250  $\mu\text{m}$  gaps between the rings. The coil design mask is then centered on the photoresist covering the  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  film on the front side of the substrate and exposed to ultraviolet light.  
15 The coil design mask is then centered on the photoresist covering the  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  film on the back side of the substrate and exposed to ultraviolet light. The resist is then developed on both sides of the substrate and the portion of the  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  film  
20 exposed when the resist is developed is etched away by argon beam etching. The remaining photoresist layer is then removed by an oxygen plasma.

25 The result is a coil structure comprised of the single crystal  $\text{LaAlO}_3$  substrate with a high temperature superconductor  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_8$  pattern of the configuration shown in Figure 1 centered on each side of the single crystal  $\text{LaAlO}_3$  substrate. The process is repeated three times in essentially the same way to  
30 produce three coils essentially identical to the first.

A schematic diagram of the experimental set-up used to demonstrate the increase in S/N when two sensors are used, and the signals from the sensors are  
35 added coherently, is shown in Figure 2. Two of the essentially identical coils are used as one sensor 11. Sensor 11 demonstrates the use of a sensor comprised of two coupled high temperature superconductor self-

resonant planar coils. The other two essentially identical coils are used as the second sensor 12. The sensors 11 and 12 are each immersed in liquid nitrogen held in Nalgene® (Nalge Nunc International, Rochester, NY) dewars 13 and 14, respectively, and each is tuned to 3.6 MHz. An Agilent E4433B Signal Generator (Agilent Technologies, Palo Alto, CA) 15 and a transmit loop 16 are used in place of a NQR source.

The transmit loop 16 is comprised of a loop of copper wire approximately 2 cm in diameter. The transmit loop 16 is placed so that the sensors 11 and 12 are approximately equidistant from it, i.e. about 0.5 m away from each of the sensors, with the plane of the transmit loop parallel to the planes of the coils of the sensors. The transmit loop is formed by removing the outer jacket and dielectric spacer from a piece of 0.080 inch (2 mm) coax cable 17. The transmit loop is formed by bending the inner conductor into a circle and soldering it to the outer jacket of the coax cable just outside the point where the jacket and dielectric are removed. The signal generator provided a -10dBm signal at a frequency of 3.6 MHz. Two pick-up coils 18 and 19, each comprised of a loop of copper wire, are placed about 1 inch (2.5 cm) away from the sensors 11 and 12, respectively, with the planes of the pick-up coils parallel to the planes of the coils of the sensors. Each of the pick-up coils 18 and 19 is formed by removing the outer jacket and dielectric spacer from a piece of 0.080 inch (2 mm) coax cable 20 and 21, respectively. The loop is formed by bending the inner conductor into a circle and soldering it to the outer jacket of the coax cable just outside the point where the jacket and dielectric are removed. The coax cables 20 and 21 are each connected to a 93 ohm 2 m piece of RG62 cable 22 and 23, respectively, which are connected to a 50 ohm tee 24. This insured that the electrical phase along the two paths is essentially

the same. The output of the tee 24 is fed to a Spectrum Analyzer R&S FSP 25 (Rohde & Schwarz GmbH & Co., KG, Munchen, Germany).

5           The signal with only one sensor present is compared to that obtained using the two sensors. The use of the two sensors resulted in a 6 dB increase in signal. This demonstrates the increase in S/N when two sensors are used to detect a frequency and the signals  
10       from the sensors are added coherently.

          While the discussion herein is presented primarily in terms of the use of a coil as the transmit, receive or transmit and receive device, this invention is not  
15       limited thereto, and is applicable to transmit, receive and transmit and receive devices that may have a configuration other than that of a coil.

          Where an apparatus or method of this invention is  
20       stated or described as comprising, including, containing, having, being **composed of or being constituted** by certain components or steps, it is to be understood, unless the statement or description explicitly provides to the contrary, that one or more  
25       components or steps other than those explicitly stated or described may be present in the apparatus or method. In an alternative embodiment, however, the apparatus or method of this invention may be stated or described as consisting essentially of certain components or steps,  
30       in which embodiment components or steps that would materially alter the principle of operation or the distinguishing characteristics of the apparatus or method would not be present therein. In a further alternative embodiment, the apparatus or method of this  
35       invention may be stated or described as consisting of certain components or steps, in which embodiment components or steps other than those as stated would not be present therein.

Where the indefinite article "a" or "an" is used with respect to a statement or description of the presence of a component in an apparatus, or a step in a method, of this invention, it is to be understood, unless the statement or description explicitly provides to the contrary, that the use of such indefinite article does not limit the presence of the component in the apparatus, or of the step in the method, to one in number.